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EXPERIMENTAL WORKSHOP ON HIGH TEMPERATURE SUPERCONDUCTORS (11 - 22 April 1988)

BACKGROUND MATERIAL TO LECTURES

BY

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THE RESONATING VALENCE BOND STATE THEORY OF HIGH TEMPERATURE CERAMIC SUPERCONDUCTORS

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ABSTRACT

High temperature superconductivity occuring in the ceramic oxide compounds La_{2-x} Sr_x Cu O_4 and Y Ba_2 Cu_3 O_{7-y} are discussed from the point of view of the Resonating Valence Bond(RVB) state theory. The RVB state is a coherent quantum liquid of Singlet Pairs of the strongly correlated electronic system described by a simple Hubbard model. Unlike the normal fermi liquid, there are two quasi particles - holons and spinons. Holons are charge +e Bosons which undergo superconducting condensation. The spinons are neutral fermions representing spin excitations. This charge-spin decoupling indicates the non-trivial departure of RVB state from ordinary metal and BCS superconductors. The spinon-helon scattering alone can explain a wealth of experimental results. Developments in RVB theories are briefly sketched.

INTRODUCTION

The field of superconductivity started when Kammerlingh Onnes observed the zero resistance state of Hg at temperatures little below $4.2^{o}K$. Soon superconductivity was discovered in many metals and compounds. Sound theoretical understanding came from the pioneering works of London, Fröhlich, Bardeen, Landau and Ginzburg which culminated in the Bardeen-Cooper-Schrieffer(BCS) theory of superconductivity. One believed that one understands the origin of superconductivity and that getting quantitative fits between theory and experiments were only a question of details and not of principles. On the experimental side, the development was phenomenal including the discovery of many new superconducting compounds and macroscopic quantum phenomena like Josepson effect, SQUIDS etc. The search for high temperature superconductors also began. As far as theoretical understanding is concerned some of the "(old) high temperature superconductors" such as Chevral phase compounds, A-15 compounds, organic superconductors defied detailed theoretical understanding and remained as bad "actors". This reached the peak with the discovery of superconductivity in La_{2-x} Ba_x Cu O_4 by Bednorz and Muller[1] This compound and the sister compound[2] Y Ba_2 Cu_3 O_{7-y} were the worst actors. Anderson[3] found that the whole wealth of experimental results on these compounds could not be fit in a honest way to conventional BCS theory. The departures were in two fronts. The first one is that superconductivity is not due to phonon induced pairing of electrons. The second and perhaps the most

important one is that superconductivity arises not from Cooper pair condensation but from condensation of new quasiparticles of charge +e which are called holons.

The ceramic superdoncutors are, as Anderson calls them, strange insulators, strange metals and strange superconductors. First let me enumerate all the strange properties of these ceramic compounds. The scale of superconducting T_c is large $\sim 94^{\circ}K$. There are indications of unstable superconductivity even at room and higher temperatures. The superconductor - normal metal tunnelling is anamolous. There is a strong ultrasonic attenuation and velocity of sound anamoly. The infrared absorption is very different from the BCS compounds. Wide discrepancies are there in the gap measurements obtained from different experiments such as tunnelling and infrared absorption.

The compound above T_c exhibits perfectly linear temperature dependent resistivity upto about 700° K. The carriers are holes, as measured from Hall and thermopower measurements - a simple band structure calculation shows that the charge carriers are electrons. There are other tetragonal to orthorhombic structural transitions which causes additional confusion to many theorists.

The insulator La_2 Cu 0_4 was well known for many years and pioneering work has come from Rao, Ganguly and collaborators[4] from our country. The copper atoms carry spin $\frac{1}{2}$ magnetic moments and they have antiferromagnetic interactions. But one did not see antiferromagnetic(AFM) long range order and the susceptibility was like

that of a Pauli paramagnet down to the lowest temperatures. Purified sample[8] did show AFM order indicating that AFM order was very sensitive to impurities.[5] AFM long range order has also been seen[6] in the Y Ba_2 Cu_3 $0_{6.25}$. The inelastic neutron scattering[7] in La_2 Cu 0_{4-y} has shown a clear indication for the presence of a quantum spin liquid (which is called a Resonating Valence Bond state or singlet liquid state).

The remarkable fact is the vicinity of the insulating phase to superconducting phase. At very low temperatures the system directly go from an insulator to a superconductor.

CRYSTAL AND ELECTRONIC STRUCTURE

We believe[8] that the copper oxides planes in the Lanthanum and 123 compound are responsible for superconductivity. Hence we will only concentrate on the La_2 Cu 0_4 compound in what follows. La_2 Cu 0_4 is isostructural to K_2 Ni F_4.(Figure 1). The Copper atoms are surrounded by an octohedra of oxygen atoms. These octohedra share corners and form a planar square network. These planes are well separated electronically. Thus if one looks at the copper plane it is a simple square lattice of copper atoms. There is an oxygen atom between every neighbouring copper atoms (figure 2). The copper oxygen distance in the plane is $\sim$ 1.9 A and the same in the vertical direction is $\sim$ 2.4 A. The copper planes are well separated by about 6 A.

La_2 Cu 0_4 is partly ionic and partly covalent in the following sense. There is complete charge transfer of 3 electrons from La atoms. Even though Cu gives up 2 electrons and becomes Cu^{2+}, there is a strong covalent binding between the copper $d_{x^2-y^2}$ orbital and the oxygen 2p orbitals. A well separated Cu^{2+} ion will be in the electronic configuration $3d^9$ - that is one hole in the d shell. The 3-d level, owing to the covalent bonding and the crystal fields from the oxygen octahedra, is split as shown in figure (3). The splitting of the top two levels (E_γ) arises because the octahedra is elongated in the Z-direction. There is enough experimental evidences which show that this large distortion dominantly arises from packing and electrostatic considerations rather than from John-Teller effect.

Thus in the insulator we have one lone electron in the oxygen $d_{x^2-y^2}$ level. The rest of the levels are far below the fermi level. The $d_{x^2-y^2}$ level hybridizes strongly with four of the oxygen 2-p orbitals pointing towards the copper atom (figure 4). As far as transport and magnetic propertice are concerned these orbitals (2 oxygen 2p orbitals and one copper $d_{x^2-y^2}$ orbital per unit cell in the plane, figure 2) alone seems to play important role.

Thus we have a simple tight binding model made up of oxygen-2p and copper $d_{x^2-y^2}$ orbitals. In the insulator there are 5 electrons per unit cell. This tight binding model in which the only nonzero hopping matrix element t_1 is between the copper orbital (of energy ϵ_1) and the neighbouring oxygen orbital (of energy ϵ_2) is easily solved to get

the dispersion relation

$$\epsilon_{k\pm} = -\frac{(\epsilon_1-\epsilon_2)}{2} \pm \frac{1}{2}\left[(\epsilon_1-\epsilon_2)^2+8t^2(1+\cos k_x a+\cos k_y a)\right]^{1/2}$$

$$\epsilon_{ko} = 0$$

The subscript $\pm$ and 0 denote antibonding, bonding and non-bonding band respectively. The corresponding dispersion is schematically shown in figure(3). The bottom band is the antibonding band, middle one is the bonding band and the top one is the antibonding band. We have three orbitals per unit cell and hence we have 3 bands. Since we have 5 electrons per unit cell, the fermi level lies in the middle of the antibonding band (figure 5). Anderson[3] has argued convincingly that the top band, namely the antibonding band, is sufficient to understand the magnetic, transport and statistical mechanical properties of this insulator. We will consider a simple tight binding model with nearest neighbour hopping between Wannier orbitals centred around the Cu sites. This Wannier orbital has the same x^2-y^2 symmetry as the $d_{x^2-y^2}$ orbital but has large oxygen 2p-orbital components as well. This tight binding model will approximately reproduce the antibonding band of our original tight binding model. In the case of La_2 Cu 0_4 this model contains one electron per site, the rest of the four electrons being there in the bonding and non-bonding bands.

Pure La_2Cu 0_4 with one electron per Wannier orbital in the above band is experimentally observed to be an insulator. When a La atom is replaced by Ba or Sr atom, it donates only 2 electrons.

Now one of the oxygen atoms receives one electron instead of two. Oxygen being strongly electronegative grabs one more electron from the copper atoms, making one of the Cu^{2+} into nominally Cu^{3+}. That is one electron from our tight binding band has been removed. Thus, when the doping fraction is x that is in $La_{2-x}\ Ba_x\ Cu\ O_4$, a fraction x of electrons have been removed. Experimentally, $La_{2-x}\ Ba_x\ Cu\ O_4$ remains an insulator upto $x \sim 0.05$ at very low temperatures and then it starts superconducting. The phenomena of removal of electrons from Cu O system by the addition of Sr or Ba will be called doping. Experimentally T_c is a function of x and T_c is maximum when $x \sim 0.15$.

One wants to understand how the insulator arises and then how it becomes a superconductor on doping.

There are magnetic and structural phase transitions that takes place as a function of doping. This is shown in the T-x phase diagram in figure (6).

MOTT INSULATOR AND MAGNETISM

The tight binding model that we considered in the last section is half-filled and will be a metal according to a simple band theory. But experimentally it is found to be a good insulator with an energy gap $\sim$ 2 eV. Such a band metal can become an insulator if a fermi surface instability due to the perfect nesting of the simple tight binding band in 2-dimension occurs resulting in a charge density wave or spin density wave. There is no experimental evidence for both. Thus one

should look for an alternate explanation for it to be an insulator. This takes us to the physics of Mott insulators.

Oxides[10] such as NiO, MnO and several antiferromagnetic insulators will be conducting metals according to simple band picture. Such a picture fails when the energy scale of electron-electron repulsion becomes large compared to the tight binding band width. That is when we have one electron per orbital, the strong electron repulsion prevents electrons from gaining the delocalisation energy by localizing one electron per orbital. When we take into account screening, the dominant electrostatic interaction is the onsite coutomb repulsion (the Hubbard U). This is typically $\sim$ 5 to 10 eV in transition metal oxides. Since the electrons hop between transition metal atoms, only through the oxygen binding orbitals, the hopping integral(t) is low $\sim$ 0.5 eV. Hence, delocalisation energy of electrons coordination number(z) x 0.5 eV $\sim$ 2 to 3 eV. This is small compared to U. This forces the electrons into a Mott insulator phase.

Any excitations involving charge transfer needs an energy $\sim$ U-zt. This is called the Mott-Hubbard gap. This gap does not arise because of any periodic (self consistant or external) potential as in a band theory. It is a genuine many body effect which escapes explanation even in terms of very 'sophisticated' density functional band theories. The Hamiltonian governing such a system is given by

The above Hamiltonian is customarily called the Hubbard Hamiltonian. Here C_i^+ creates an electron at the orbital at site i, t is the hopping integral, U is the onsite repulsion between two electrons and μ is the chemical potential.

One believes that the Hubbard Hamiltonian describes a Mott insulator when U/t is greater than a critical value in 2 and 3 dimensions. Even though the charge excitations have a finite gap, the spin fluctuations have no gap. The spin fluctuations which are the low lying states are described by an effective Hamiltonian usually referred to as the Heisenberg exchange Hamiltonian:

$$H = J \sum (\vec{S}_i \cdot \vec{S}_j - \frac{1}{4})$$

Where $J = \frac{2t^2}{U}$ and $\vec{S}$ are the Pauli spin operators. In terms of the original electron operators this Hamiltonian has a simple form

$$H = \frac{-J}{2} \sum b_{ij}^+ b_{ij} \qquad (3)$$

$$\text{Where } b_{ij} = (C_i^+ C_j^+ - C_i^+ C_j^+) \qquad (4)$$

The exchange described by equation (2) is indeed the Kramers - Anderson's Superexchange[11] well known in insulating oxides. It has been rigorously established that spin - $\frac{1}{2}$ Heisenberg antiferromagnet has long range order at low temperatures in 3 and higher dimensions. In one dimension Quantum fluctuations destroy long range order converting it into a Resonating Valence Bond(RVB) state. RVB state is a quantum liquid

of singlet pairs. A good operational definition of RVB state is as follows.

$$| \text{RVB} \rangle = P_G \left(\sum a_{ij} b_{ij}^+ \right)^{\frac{N}{2}} | 0 \rangle \qquad (5)$$

$$\text{with } P_G = \prod_i (1 - \hat{n}_i \hat{n}_i) \qquad (6)$$

$$\text{and } a_{ij} = a(| i - j |), \qquad (7)$$

The operator $\sum a_{ij} b_{ij}^+$ creates a pair of electrons in a singlet state and with centre of mass momentum equal to zero. The function $a(|i-j|)$ is a decreasing function of separation $|i-j|$. In the well studied nearest neighbour bond RVB state,[12] $a(|i-j|) = 0$ for $|i-j|$ greater than the nearest neighbour distance. The Gutzwiller projector P_G projects out any doubly occupied state arising from overlapping singlets. The fact that singlets have been created in zero momentum state gives phase coherence among various singlet configuration appearing in the RVB wave function. As Anderson[14] noticed, this phase coherence is exactly the one that appears in a superconducting BCS state. What prevents an RVB state from becoming superconductor is the absence of any real double occupancy or zero occupancy. Another way of saying is that it is a BCS state with zero electronic compressibility.

Anderson[12] observed that oxides which are $S = \frac{1}{2}$ Heisenberg antiferromagnets rarely have long range magnetic order. Their low temperature magnetic properties are often anomalous. From this he concluded that

perhaps AFM order is fragile and could easily be destroyed. He constructed in 1973 the example of 2-d triangular lattice where low dimension and geometric frustration resulted in a RVB state.

Much latter Anderson, Baskaran, Zou and Hsu(ABZH)[15] argued that when less than 1% of holes are introduced in an antiferromagnetic Mott insulator, the long range order is destroyed and the insulator is converted into a RVB liquid. The basic argument is similar to Nagaoka's theorem.[16] The hole wants to delocalize and gain maximum binding energy $\sim$ tz. In an AFM state the delocalisation energy gain is not maximum.[17] This is possible only when the bulk magnetic state readjusts itself. This rearrangement costs an energy J. For $\frac{U}{t}$ very large this rearrangement is into a ferromagnetic state (Nagaoka theorem). When $\frac{U}{zt} \sim 1$ to 10, which is the experimentally relevant range, the rearrangement is into an RVB liquid. The transition occurs when the energy loss of the bulk magnetic energy per site ΔE becomes comparable to hole delocalisation energy per site $\sim tz$.

All the above agreement shows that Mott insulator can be easily converted into a quantum liquid of singlet pairs with very good phase coherence among the various singlet configurations. We will argue that such a state is a potential high temperature superconductor.

SUPERCONDUCTIVITY OF THE DOPPED MOTT INSULATOR

Very soon after the discovery of high temperature superconductivity became known to the world, Anderson[14] gave a Hubbard model description and provided a qualitative RVB scenerio of superconductivity. His basic idea was that the singlet pairs in the RVB insulator are like cooper pairs.

In the Mott insulator they are neutral owing to the absence of real charge density fluctuation in an insulator. When Ba is added and electrons are removed from the Mott insulator, a fraction of the pre-existing singlet pairs get charged and superconductivity results. He also pointed out that the ground state wave function is

$$| \text{RVB} \rangle = P_G \left(\Sigma a_{ij} b_{ij}^{+} \right)^{\frac{N}{2}(1-x)} | 0 \qquad (8)$$

where x is the doping fraction. The above wave function, eventhough resembles the n-particle projection of a BCS wave function, has qualitatively different behaviour. Anderson also pointed out that in such a correlated fermi liquid gapless spin excitations may be still present. He argued that they are the neutral spin $\frac{1}{2}$ fermions having their own fermi surface (called pseudo fermi-surface) contributing to the experimentally observed Pauli like susceptibility of the insulator. The remarkable point is that he conjectured that this pseudo fermi surface could remain intact even after doping resulting in a linear low T specific heat of the superconductor.

A mean field theory which explicitly brought out the pseudo fermi surface of the insulator and superconductivity of the doped insulator was developed by Baskaran, Anderson and Zou(BZA).[18] One starts with the effective Hamiltonian of the doped insulator (for less than half filled band)

$$H_{eff} = -t \; \Sigma (C_i \, C_j + h.c)$$

$$-\frac{J}{2} \; \Sigma b_{ij}^{+} \, b_{ij} - \frac{J}{2} \; \Sigma \; b_{ij}^{+} \, b_{jk} \qquad (9)$$

With the constraint $n_i + n_i = 1$ for every site. A BCS type of factorisation $b^{\dagger}_{ij} b_{ij} \sim \langle b_{ij} \rangle b_{ij} + h.c.$ and relaxing the constraint $n_i + n_i = 1$ gave several interesting results and insights into the problem.

An observation by Kivelson, Rokshar and Sethna[19] brought out a nontrivial departure from conventional BCS theory. They argued, by taking analogy from Peierls and spin-Peierl's insulator, that in our doped valence bond system the charge carriers are 'holons' having a charge of +e. They behave like bosons and superconductivity results from the Bose condensation of these holons. A holon is essentially an empty site with the rest of the electrons singlet bonded and resonating among various valence bond configuration in a coherent way. When such an empty site is filled with one electron we get a spinon which is a neutral fermion. A spinon is essentially an unpaired spin in a sea of resonating singlet pairs. Thus the holons and spinons are quasiparticles of an RVB superconductor.

At low temperatures the holons have perfect phase coherence leading to superconductivity. The effective mass of these holons $\frac{h^2}{2a^2 t}$ where a is the lattice parameter. In an ideal Bose system this will set the scale for the superconducting transition temperature, which will be $\sim 5000^{\circ}$ K! However, the physics in our problem is more complicated. At finite temperatures, spinon pairs are thermally librated from the RVB sea of singlet pairs. They associate themselves with the holons thereby reducing their Boson like phase coherence. The phase coherence is completely lost when $k_B T \sim J$, at which temperature most of the singlet pairs have been broken. This temperature sets the scale for the superconducting transition temperature.

The holon condensation has, as pointed by KRS, dramatic consequences. The important one among them is the magnetic field flux quantization in units of $\frac{hc}{e}$ and not $\frac{hc}{2e}$ as in the conventional BCS superconductors. Recent experiments have shown that indeed the flux quantisation is $\frac{hc}{2e}$. That this does not contradic the RVB theory of superconductivity has been explained by Wheateley, Hsu and Anderson(WHA)[20] in the following way. In La_2 Cu 0_4 as well as the 1 2 3 compound the copper oxide planes are well separated and hence electronic conduction is highly anisotropic. The holon condensation in the 2-d plane does bring in superconducting Kosterlitz-Thouless type of correlation within the plans. But there is no true off-Diagonal Long Range Order(ODLRO). Due to the strong singlet correlations within the plane single electron tunnelling is less probable than tunnelling of a pair electrons. This dominant pair tunnelling establishes a 3-dimensional condensate of 2e rather than e. This is consistant with the experimental results on flux quantisation and Josephson effect. WHA have also developed a simple mean field theory to study the consequences of the above picture.

The developments in the theory of RVB superconductivity were varied and were apparently different looking. Soon after the BZA mean field theory KRS brought the picture of charge +e holon condensation. Some local discrete symmetries in the Mott insulators and in the BZA mean field theory was used to explain(ABZH) the universal structural phase transitions (named "twitch" transition) in the ceramic compounds.

Then certain local continuous symmetries were recognized and this resulted in Baskaran and Anderson's Gauge (BAG) theory.[21] Soon Anderson discovered an SU(2) local symmetry[3] and this was formalised recently by Affleck, Zou, Hsu and Anderson(AZHA).[22] The BAG theory argued that the long wave length and long time scale behaviour of the Mott insulator is governed by a fixed point Hamiltonian which has a local U(1) symmetry. AZHA showed that infact it is an SU(2) local symmetry.

Soon after KRS's suggestions, Zou and Anderson(ZA) did a Slave Boson mean field theory bringing out the holon condensation explicitly. The approximations seemed to violate certain local conservation laws. Recently I have developed a theory which combines the BAG theory and ZA theory and provides a framework for further developments. It also gives an unified picture of the previous developments. The holon condensation is brought out by this theory through an Anderson-Higgs type of mechanism in a natural way.

RELATION OF RVB THEORIES TO EXPERIMENTS

After about an year of developments we realize that all the anomalous properties of metal superconductor and insulator results from the so called charge- spin decoupling in a strongly correlated electronic system. This realisation alone can explain a wealth of experimental results in a simple and natural way. In a correlated RVB metal the charged quasi particles are holes without any spin. The neutral quasi particles are spinons which are fermions. This is different from the

quasiparticles of conventional fermi liquid which are fermions. There is only one type of quasi particle which carry both the spin and the charge in the normal fermi liquid.

The presence of spinons and holons and the strong scattering between them has been used to explain the linear resistivity-temperature dependence of anisotropic resistivities and superconductor - metal tunnelling characteristics. The charged bosons scatter against the spinons. The spinons are excitations out of a pseudo fermi sea. The density of these quasi particles is proportional to T for $T < J$. Hence the life time of quasiparticles is $\sim T$. This explains the linear resistivity.[24]

Careful tunnelling measurements[25] on thin films of the ceramic superconductor show a gapless conductance VS. voltage character. Anderson and Zou[24] have also explained this as follows. Tunnelling across a junction occurs by real electron tunnelling and not by holon tunnelling. Thus the holon, when it reaches a junction, has to wait for a thermally produced spinon to come and convert it into a real fermion. This beautiful and simple fact explains satisfactorily the tunnelling characteristics as well as the strong temperature dependent resistivity in the direction perpendicular to the Cu O planes.

The scale of transition temperature follows easily from the RVB theories. And indeed it gives hope for room temperature superconductors. The best candidates for room and higher temperature superconductivity are 3-dimensional RVB systems. At present the 3-d transition temperature is determined by the dopant concentration (x) and by the interplanar Josephson type of coupling.

In 3-d RVB system this coupling will be stronger and we can hope to reach a $T_c \sim J$.

The experimental discovery of high temperature superconductivity has opened new chapters in the physics of highly correlated electronic systems. Apart from the fascinating high temperature superconductivity many other things happen. There are new type of quasi particles. I have recently argued that infact there are two type of metals - the conventional fermi-liquid metal and the RVB metal atleast at T = O. RVB metal occurs for moderate doping and U/t greater than the critical value. They are governed by two different fixed points. The interaction of the spinons and holons becomes a theory involving collective SU(2) gauge fields. The structural phase transitions is intimately related to the RVB fluctuations.

Until recently most of the RVB development have been rather qualitative owing to the emergence of the rich physics. Soon one can hope to sea quantitative and new developments in this rich field. I have tried to provide only a flavour of RVB phase and it is very clear a lot more can be done.

Figure Captions:

1) $K_2 Ni F_4$ type crystal structure of $La_2 Cu O_4$
2) The $Cu O_2$ planes found in $La_2 Cu O_4$ as well as $Y Ba_2 Cu_3 O_4$
3) The ligand field split levels of Cu 3d levels in the octahedral environment
4) The copper $d_{x^2-y^2}$ and oxygen 2-p orbitals involved in the relevant electronic band where the fermi level lies.
5) The schematic picture of the bonding, non-bonding and antibonding band
6) The T-x phase diagram showing the magnetic, structural and superconducting phase transitions.

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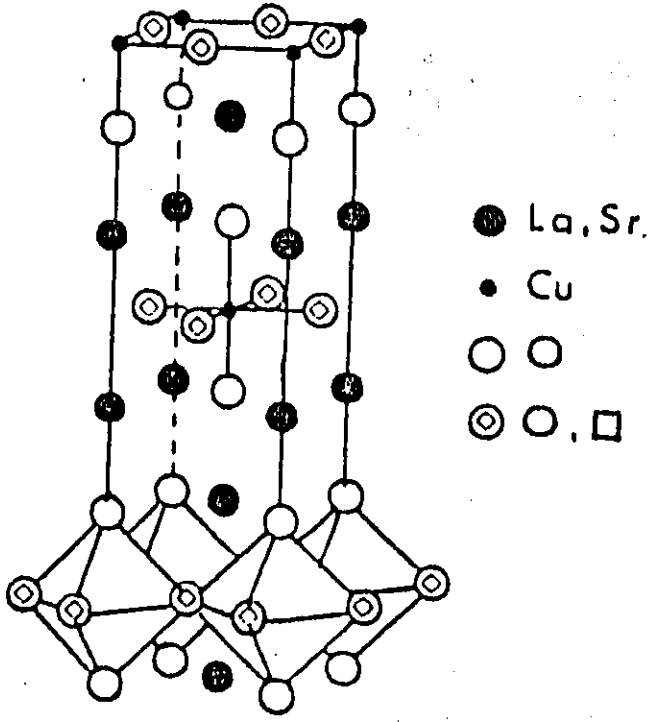

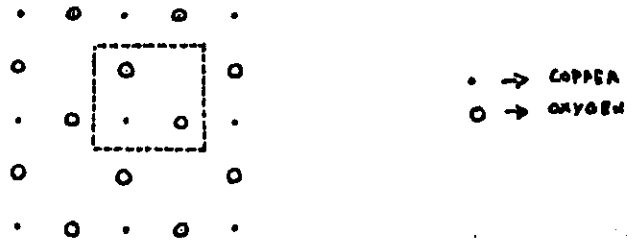

FIGURE 2

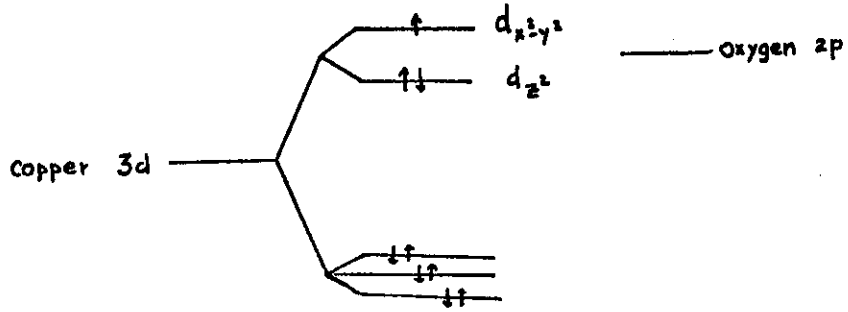

FIGURE 3

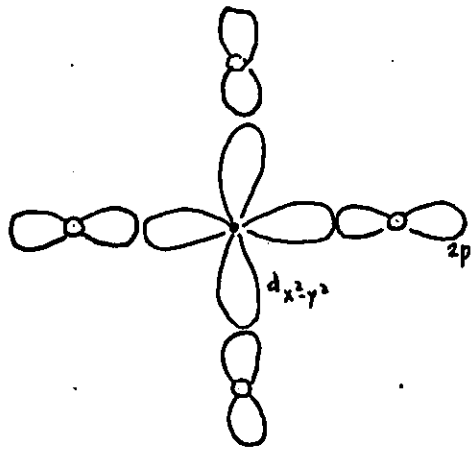

FIGURE 4

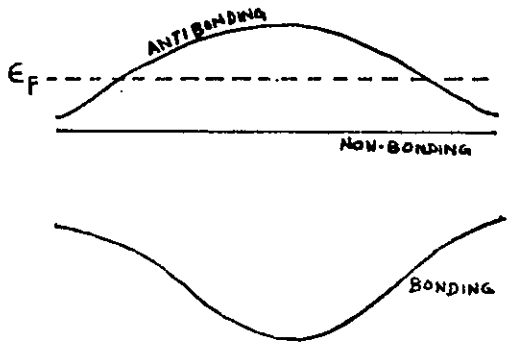

FIGURE 5

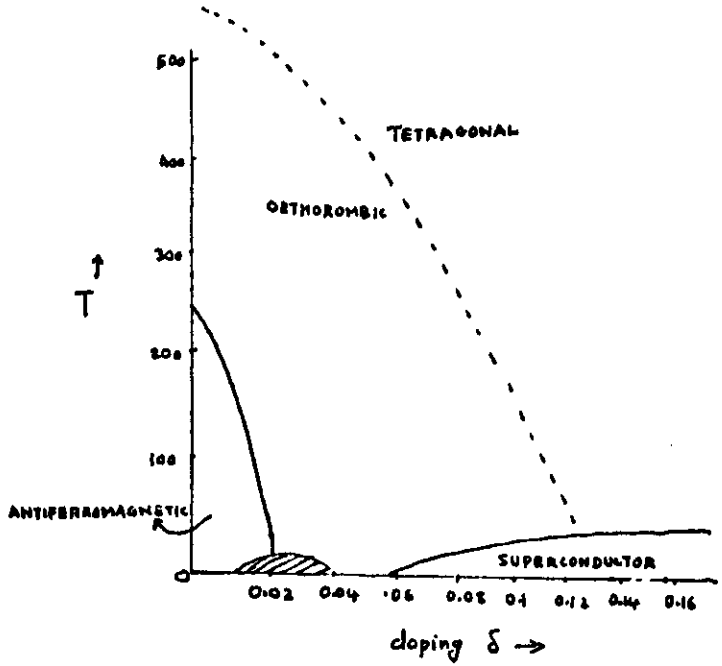

FIGURE - 6.